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| (21) International Application Number: PCT/US95/12801 (22) International Filing Date: 12 October 1995 (12.10.95) (30) Priority Data: 08/322,883 13 October 1994 (13.10.94) US (71) Applicant: AKZO NOBEL N.V. [NL/NL]; Velperweg 76, P.O. Box 9300, NL-6800 SB Arnhem (NL). (71)(72) Applicants and Inventors: MOY, Paul, Y. [US/US]; 10 Sandi Lane, Fishkill, NY 12524 (US); BRIGHT, Danielle, A. [US/US]; 21 Zabela Drive, New City, NY 10956 (US). (74) Agent: FENNELLY, Richard, P.; Akzo Nobel Inc., 7 Livingstone Avenue, Dobbs Ferry, NY 10522 (US). | | (81) Designated States: CA, CN, JP, European patent (AT, BE, CH, DE, DK, ES, FR, GB, GR, IE, IT, LU, MC, NL, PT, SE). Published <i>With international search report.</i> |
| (54) Title: POLYCARBONATE-CONTAINING POLYMERS FLAME RETARDED WITH OLIGOMERIC PHOSPHATE ESTERS (57) Abstract <p>Polycarbonate-containing compositions are flame retarded with a mixture which comprises an arylene-bridged oligomeric polyphosphate ester and an effective amount of an alkylene-bridged diphosphate compound for increased flame retardancy efficacy and improved processing characteristics.</p> <p>FOR U.S. APPLICATION IDS</p> | | |

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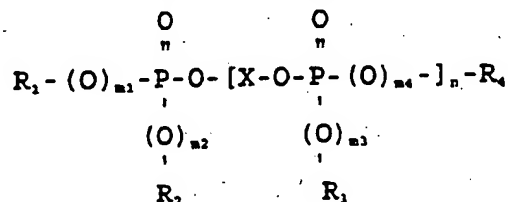
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**POLYCARBONATE-CONTAINING POLYMERS
FLAME RETARDED WITH OLIGOMERIC PHOSPHATE ESTERS**

Background of the Invention

U.S. Patent No. 5,204,394 to J.C. Gosens et al. advocates the use of arylene-bridged oligomeric polyphosphate esters in the flame retarding of polycarbonate-containing polymer compositions. An oligomeric phosphate or blend of oligomeric phosphates are disclosed by the Gosens et al. patent as having the formula



where R_1 , R_2 , R_3 , and R_4 each represent an aryl or an alkaryl group chosen independently of each other and wherein X is an arylene group, m_1 , m_2 , m_3 , and m_4 , each independently of each other are 0 or 1 and wherein $n=1, 2, 3, 4$, or 5 or wherein, in the case of a blend of phosphates, n has an average value between 1 and 5.

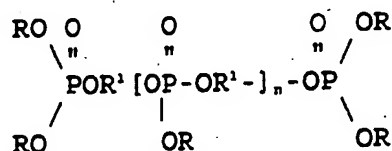
Summary of the Invention

It has now been found that a mixture of an alkylene-bridged diphosphate compound and the foregoing type of arylene-bridged oligomeric polyphosphate ester is an effective flame retardant agent in the aforementioned types of polycarbonate-containing polymer compositions. It has been found that there is increased flame retardancy efficacy and improved processing characteristics for the mixture in such polymers.

Description of Preferred Embodiments

The polycarbonate-containing resins and the arylene-bridged oligomeric phosphate esters to which the present invention is applicable are described in the aforementioned U.S. Patent No. 5, 204,394 which is incorporated herein by reference in regard to such teaching.

The terminology "alkylene-bridged diphosphate compound" as used herein is to be understood to encompass monomeric and low oligomeric species of the formula



Where R is aryl, such as unsubstituted phenyl, n is a number ranging from 0 to about 5, and Rⁱ is alkylene of from 1 to 8 carbon atoms.

The present invention contemplates using a mixture comprising the arylene-bridged phosphate compositions and the alkylene-bridged compounds in a ratio of from about 1:2 to about 2:1 on a weight basis in the polycarbonate-containing compositions. Generally speaking, the amount of such mixture used in the polycarbonate-containing compositions will vary from about 1 % to about 25 % on the weight of the polycarbonate-containing composition.

The present invention is illustrated by the Examples which follow.

EXAMPLES 1-5

These Examples set forth cone calorimetric data (35.0 kW/m² irradiance) on neat PC/ABS, neat resorcinol diphosphate (RDP) and blends of RDP and neopentylglycol diphosphate at various weight amounts.

The polymers composites were preblended to a five to one (PC/ABS) ratio and extruded in a twin screw extruder to a homogeneous blend. The phosphate flame retardant mixtures ("FR" in the Table given below) were preblended in the prescribed ratios before addition to the composites. The incorporation of the flame retardants was accomplished by metering the liquid components into the melt stream of the extruder through a side port injector. All flame retardant composites are formulated to contain a small amount of powdered TEFLON fluoropolymer antidrip agent.

TABLE 1

| | | %P | Peak Heat Release | Total Heat | Effective Heat of Combustion |
|----|-----------------|------|----------------------|------------|---------------------------------|
| | | | (kW/m ²) | (KJ) | (MJ/kg) |
| 25 | <u>FR USED</u> | | | | |
| | NONE | - | 201 | 32.12 | 3.48 |
| | RDP (Neat) | 0.91 | 69 | 17.79 | 2.04 |
| | RDP/NPGDP (2/1) | 0.92 | 62 | 18.83 | 2.02 |
| 30 | RDP/NPGDP (1/1) | 0.71 | 52 | 14.66 | 1.61 |
| | RDP/NPGDP (1/2) | 0.80 | 43 | 16.04 | 1.85 |

EXAMPLES 6-10

These Examples show the melt flow viscosity data (ASTM-1238, Condition "I", 230°C, 3.8 kg) for the blends tested in each of Examples 1-5, above.

FR Sample (from Ex. 1-5) g/10 minutes

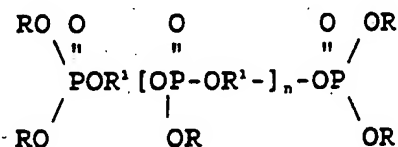
| | | |
|----|-----------------|------|
| | NONE | 13.0 |
| 10 | RDP (Neat) | 16.9 |
| | RDP/NPGDP (2/1) | 18.8 |
| | RDP/NPGDP (1/1) | 15.3 |
| | RDP/NPGDP (1/2) | 17.3 |

15 The foregoing data is presented for purposes of illustrating certain embodiments of the present invention and, for that reason, should not be construed in a limiting sense. The scope of protection sought is set forth in the claims which follow.

We Claim:

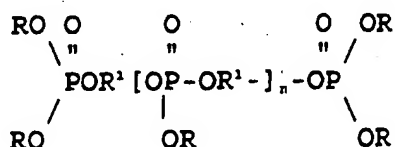
1. A flame retarded polycarbonate-containing composition which comprises a mixture comprising (1) an arylene-bridged oligomeric polyphosphate ester and (2) an effective amount of an alkylene-bridged diphosphate compound for increased flame retardancy efficacy and improved processing characteristics.

2. A composition as claimed in Claim 1 wherein the alkylene-bridged diphosphate compound is of the formula



wherein R is aryl, n is a number ranging from 0 to about 5, and R¹ is alkylene of from 1 to 8 carbon atoms.

3. A composition as claimed in Claim 1 wherein the alkylene-bridged diphosphate compound is of the formula



where R is unsubstituted phenyl, n is a number ranging from 0 to about 5, and R¹ is alkylene of from 1 to 8 carbon atoms and the arylene-bridged ester and the alkylene-bridged compound are in a ratio of from about 1:2 to about 2:1 on a weight basis.

4. A composition as claimed in Claim 1 wherein the amount of such mixture used in the polycarbonate-containing compositions will vary from about 1 % to about 25 % on the weight of the polycarbonate-containing composition.

5. A composition as claimed in Claim 2 wherein the amount of such mixture used in the polycarbonate-containing compositions will vary from about 1 % to about 25 % on the weight of the polycarbonate-containing composition.

6. A composition as claimed in Claim 3 wherein the amount of such mixture used in the polycarbonate-containing compositions will vary from about 1 % to about 25 % on the weight of the polycarbonate-containing composition.

7. A composition as claimed in Claim 1 wherein the arylene-bridged oligomeric polyphosphate ester is resorcinol diphosphate and the alkylene-bridged diphosphate compound is neopentylglycol diphosphate.

8. A composition as claimed in Claim 3 wherein the arylene-bridged oligomeric polyphosphate ester is resorcinol diphosphate and the alkylene-bridged diphosphate compound is neopentylglycol diphosphate.

9. A composition as claimed in Claim 5 wherein the arylene-bridged oligomeric polyphosphate ester is resorcinol diphosphate and the alkylene-bridged diphosphate compound is neopentylglycol diphosphate.

INTERNATIONAL SEARCH REPORT

International application No.
PCT/US95/12801

A. CLASSIFICATION OF SUBJECT MATTER

IPC(6) :C08K 5/523

US CL :524/127

According to International Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)

U.S. : 524/127

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practicable, search terms used)

C. DOCUMENTS CONSIDERED TO BE RELEVANT

| Category* | Citation of document, with indication, where appropriate, of the relevant passages | Relevant to claim No. |
|-----------|---|-----------------------|
| Y | US, A, 5,204,394 (GOSENS ET AL.) 20 April 1993, see column 2, lines 5-41. | 1-9 |
| Y | US, A, 3,869,526 (COMBEY ET AL.) 04 March 1975, see column 1, lines 1-29 and column 3, lines 29-50. | 1-9 |
| Y | JP, A, 59-202,240 (DAIHACHI KAGAKU KOG KK) 16 November 1984, see the abstract | 1-9 |
| Y | JP, A, 59-45,351 (ADEKA-ARGUS CHEM KK) 14 March 1984, see the abstract. | 1-9 |



Further documents are listed in the continuation of Box C.



See patent family annex.

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